

Structural and optical quality of binary photonic crystal heterostructures fabricated by the modified self-assembly method

G.Q. Liu*, Y.H. Ji, Y.Y. Nie, Y. Chen and H.H. Hu

*College of Physics and Communication Electronics, Jiangxi Normal University,
Nanchang, Jiangxi 330022, People's Republic of China*

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Photonic crystal heterostructures constituting of two photonic crystals with different lattice constants are fabricated using the modified self-assembly method and their structural and optical properties are investigated. The results show that these photonic crystal heterostructures of high quality possess deep photonic band gaps and steep photonic band edges in their transmission spectra. Deep double photonic band gaps, steep photonic band edges and high transmittance in the pass band show good ordering of the heterostructure and may offer a probability for studying late-model ultra-fast all-optical switches.

Keywords: photonic crystal heterostructures; double photonic band gaps; self-assembly

1. Introduction

Photonic crystal (PC) heterostructures exhibit many optical properties which resemble the superposition of the properties of each individual crystal [1,2], afford new flexibility and functionality for engineered photonic behavior in practical devices, such as the low-loss PC waveguide, record resonator quality factors and high-efficiency add-drop filters etc. [3,4]. PC heterostructures achieved by combining two PCs with different lattice constants have been demonstrated [5–9] and their optical properties were analyzed from the reflection or transmission modes. High quality PC heterostructures with deep double photonic band gaps (PBGs) and steep photonic band edges (PBEs) may offer a strategy for studying late-model ultra-fast all-optical switches due to the nonlinear optical effects of polymer. However, it is still a challenge to achieve high quality PC heterostructures with deep PBGs and steep PBEs before practical applications. To fabricate high quality PC heterostructures with deep PBGs and steep PBEs, the crucial approaches are to improve the quality of the constituent PCs and to modify the roughness at the interface. The former mainly relies on the technique employed to fabricate the 3D PCs, the latter is closely related to the elimination of lattice mismatch occurring at the interface which can be largely eliminated when bigger particles are deposited on top of small particles [9].

Recently, we have established a modified self-assembly system and assembled high quality PCs with deep PBGs and steep PBEs from polystyrene (PS) colloidal suspensions successfully [10]. This method provides a basis for fabricating PC heterostructures with high quality. In this paper, we fabricate two types of binary PC heterostructures of small particles directly deposited on a glass substrate followed by bigger particles using the modified self-assembly method. The optical properties of the resultant binary heterostructures are mainly investigated from the transmission spectra. This is because the transmission scans the whole thickness of the heterostructure. Furthermore, the characteristics (e.g. PBG depth and PBE slope) of the transmission spectra closely contact with the roughness of the interface between the constituent PCs and the quality of the heterostructures.

2. Sample fabrication and measurement

Three types of PS spheres (Duke Scientific Corp.) with diameters 260, 350 and 360 nm were used to generate PC heterostructures. The dispersion is less than 3% and their refractive index (n) is ~ 1.59 . The aqueous solution of PS spheres was diluted to 0.5 wt% by de-ionized water. A glass substrate ($n \approx 1.46$) with a dimension of $2 \times 1.8 \text{ cm}^2$ was cleared with ethanol

*Corresponding author. Email: liougq@yahoo.com.cn

absolute, dried in an oven and then fixed vertically into the PS suspension contained in the deposition vial. The vial was sealed and connected to the vacuum pump through a pipe and a sealed glass service, as shown in Figure 1. The pressure in the growth chamber was controlled by the vacuum pump together with the small valve attached to the pipe and the temperature was controlled by the isothermal bath. When the vacuum pump was started, the evaporated steam was driven to flow quickly through the pipe and into the sealed glass service. A stable growth condition, beneficial to fabricate high quality PCs, was generated by controlling the speed of the evaporation of the water vapor. The evaporation speed was adjusted by the pressure and temperature. Influenced by the surface tension and capillary force PS colloidal spheres will deposit on the substrate and form high quality PCs. Then, the sample was air-dried for ~ 20 h and then dipped again into another PS suspension with different particle sizes to form the next PC. The thickness of each PC can be easily tuned by adjusting the concentration of the PS solution. The PCs self-assembled in this way possess only a directional band gap (pseudogap) along the [111] direction because of the low dielectric contrast with respect to air. Although a complete band gap is not available in these PCs, they still attract much attention due to their unique advantages over other PCs. 3D PCs self-assembled from PS spheres have been successfully employed to construct ultra-fast all-optical switches due to the large nonlinear optical effect of the polymer [11,12].

Two types of PC heterostructures are fabricated. They are denoted as AB and CB structure, respectively. Here, A, B and C denote the crystals composed of PS spheres of 260, 360 and 350 nm in diameter, respectively. We describe the multilayer PC heterostructures pattern by listing the letter from bottom to top according to the deposition sequence of particles. For example, the AB structure denotes the

heterostructure formed by depositing crystal A on the surface of the glass substrate, followed by crystal B. Since the second PC was composed of PS spheres with different sizes, a different growth condition was needed in order to achieve a high quality PC. Therefore, the pressure in the vial for fabricating crystals A, C, and B were set at 45, 42, and 40 mmHg, respectively. The temperature during the fabrication of crystals A, C and B was kept at 35, 37, and 39°C, respectively.

Optical characterization of these samples was measured using a spectrophotometer with a halogen lamp as the light source, operating both in reflection and transmission modes. The measurement scheme of reflection and transmission modes is shown in Figure 2. The incidence light emitted from the halogen lamp was focused into the narrow slit of a grating spectroscopy and divided into monochromatic light with a wavelength range of 350–1200 nm and then focused on the sample. The dimensions of the beam irradiated on the samples was $5 \times 2 \text{ mm}^2$. The reflection spectra were probed at an angle of 10° from normal incidence with respect to the (111) plane of the PC. A photoelectricity detector was used to selectively detect light from $7.5 \times 3 \text{ mm}^2$ circular regions of the sample. The structural characterization was done with a scanning electron microscope (SEM).

3. Characteristic analysis

Figure 3(a) depicts the near-normal incidence ($\theta \sim 10^\circ$) reflection measured for the AB heterostructure formed from 360 nm particles directly deposited on top of the 260 nm particles. The thickness of the AB structure is 22 layers for crystal A and 19 layers for crystal B. The reflection spectra are probed from both the top of the samples (dashed line) and the bottom, i.e. through the glass substrate (solid line). Two peaks obtained in reflection spectra for the AB structure are consistent with the corresponding PBG of each individual crystal. Obviously, the peak at ~ 600 nm corresponding to the first order Bragg reflection from crystal A is very intense when the sample is probed through the underlying glass substrate, yet very weak when probed from the top, through crystal B. The reduction in reflectance at ~ 600 nm along the opposite direction is from 47% to 10%. A similar change in intensity of the peak at ~ 830 nm along the opposite direction is observed. It is well known that the density of imperfections in a PC can be reduced by choosing an effective technique but can not be eliminated completely. When crystal B directly crystallizes on the surface of crystal A, the increased roughness of crystal A compared to the bare substrate will introduce disorders and defects other than those inherent to the

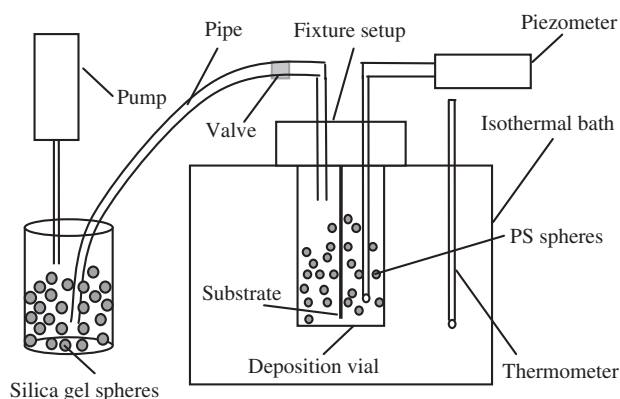


Figure 1. Systematic setup of self-assembly.

self-assembly process in crystal B. These will affect the structural and optical quality of the whole PC heterostructure. Light with some frequency propagating in the PC heterostructure will be scattered randomly, resulting in the weakened intensity of reflection light measured in a finite area. Therefore, we propose that the phenomena occurring in Figure 3(a) arise from the fact that the illumination of the AB structure from the top, the intensity of light with a wavelength of ~ 600 nm is largely scattered by the disorder in crystal B when first passing through the region of crystal B, reflected back to crystal B by crystal A with a little attenuation, and then scattered largely again by the second passing through the region of crystal B. In contrast when illuminated from the bottom, the light is directly reflected by the first region of crystal A with only a little attenuation. In addition, it is possible that the illuminated regions may be different when measuring from the top and the bottom of the sample. Therefore, the attenuation in intensity of the reflected light is attributed to the presence of impurity scattering and also to differences in the illuminated region for the two cases.

The transmission spectra measured along normal incidence from both the top (solid line) and the bottom (dashed line) of the AB structure are shown in Figure 3(b). Obviously, the difference between the transmission spectra measured in both directions is far less than that between the reflection spectra. The light directly transmitting through the sample is attenuated only once by each constituent crystal in the sample. As a result, the influence of the disorder in the sample is largely diminished. Therefore, the transmitted light propagating along only one direction, differing from the reflected light, may be the main explanation. As shown in Figure 3(b), the peaks near 830 nm in these two spectra are almost the same, indicating that the optical properties of crystal B are affected little by crystal A due to the high quality of the latter. However, in Figure 3(b), the transmission spectrum measured from the bottom of the sample possesses a shallower PBG at ~ 600 nm as compared to that measured from the top. It is because the intensity of light with a wavelength of ~ 600 nm is partially scattered by the disorder in crystal B when first passing through the region of crystal B.

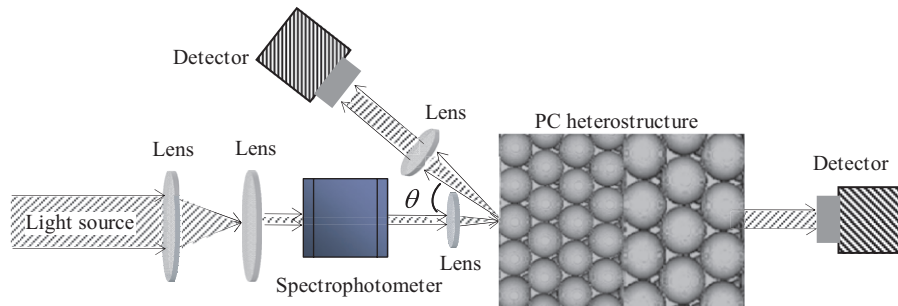


Figure 2. Sketch of the experimental setup for the measurement of the reflection and transmission modes for the PC heterostructures. (The color version of this figure is included in the online version of the journal.)

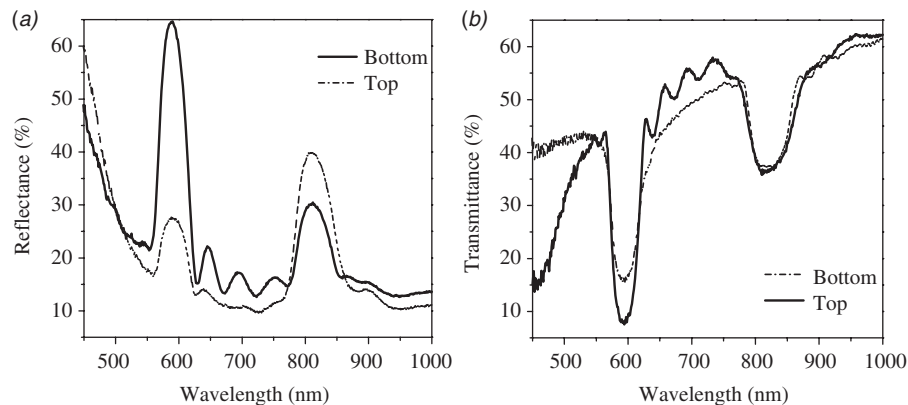


Figure 3. (a) Reflection spectra for the AB structure measured from the bottom (solid line) and the top (dashed line) of the structure. (b) Transmission spectra for the AB structure measured from the bottom (dashed line) and the top (solid line) of this structure.

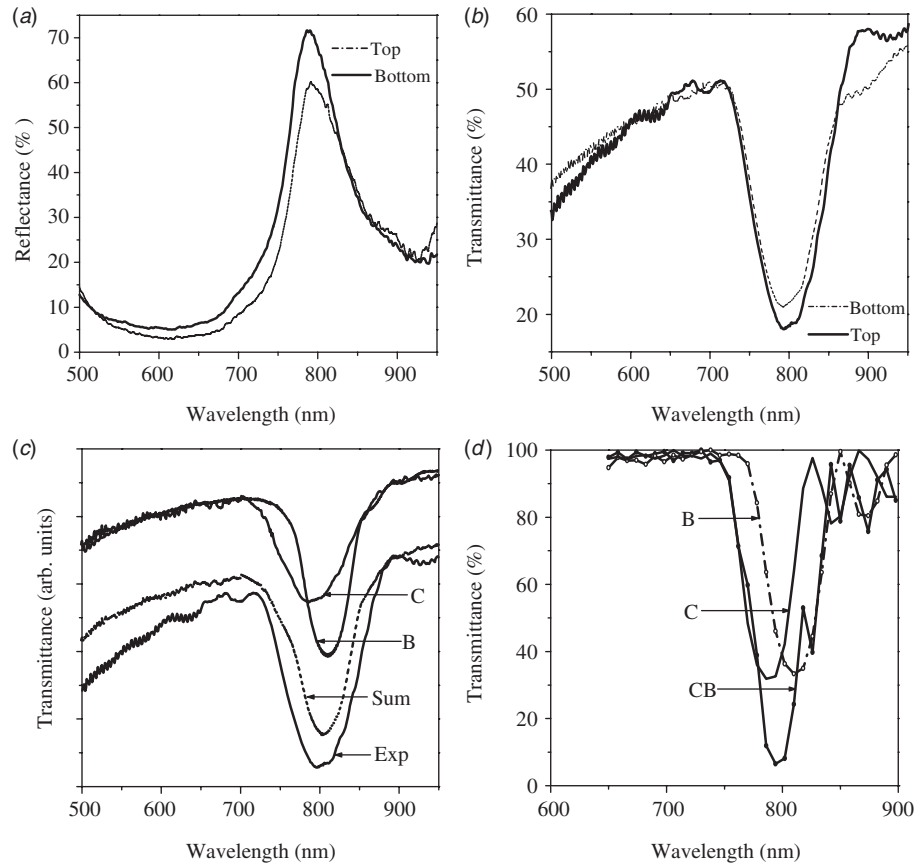


Figure 4. (a) Reflection spectra for the CB structure measured from the bottom (solid line) and the top (dashed line) of the structure. (b) Transmission spectra for the CB structure measured from the bottom (dashed line) and the top (solid line) of this structure. (c) Transmission spectra of CB structure (Exp), individual crystals C and B and the sum of these two reference spectra (Sum). (d) The simulation results of the CB structure and individual crystals C and B.

In Figure 3(b), two deep dips (PBGs) with steep PBEs are clearly observed. The transmission spectrum measured from the top of the sample is analyzed as an example. In this transmission spectrum, two deep dips (PBGs) with steep PBEs are clearly seen here. The PBG depth for crystal A is $\sim 40\%$ and for crystal B is $\sim 27\%$ and the transmittance in the pass band between the two PBGs is as large as 55% . However, as reported in [2,9], the band gap depths corresponding to crystals A and B in the AB structure are less than 13% and 18% , which are much less than those observed in our heterostructure. The transmittance in the pass band between the two band gaps (30%) in [2,9] is much lower compared with our heterostructure. In addition, in contrast to the results shown in previous work [2,5–9,13], clear F-P fringes [14,15] found in our heterostructure AB implies a high uniformity and less imperfections in our heterostructure. More importantly, the band gaps in our heterostructure possess much sharper band edges with differential transmission $4\% \text{ nm}^{-1}$ for crystal A and $2.8\% \text{ nm}^{-1}$ for crystal B. All these features indicate that a significant progress in the fabrication of PC heterostructures was achieved by using the

modified self-assembly method. Furthermore, deep PBG and steep PBE of PCs are especially important for producing optical devices based on PBE or PBG shift of tunable PBG crystals, e.g. ultra-fast all-optical switches. High quality PC heterostructures with deep double PBGs and steep PBEs may offer a probability for studying late-model ultra-fast all-optical switches due to the nonlinear optical effects of PS colloidal spheres. When we deliberately choose the frequencies of the control signals near the band edges, two signal lights with different frequencies can be simultaneously or selectively switched off through the pumping of the binary PC heterostructure with this control light.

Another PC heterostructure was formed from 360 nm (B) particles directly deposited on top of the 350 nm (C) particles, represented as CB structure. Figure 4(a) shows the reflection spectra from the top (dashed line) and the bottom (solid line) of this sample. The peak in the reflection spectrum measured from the top of this structure is slightly weaker than that measured from the bottom. As analyzed above, this difference also can be attributed to the presence of impurity and also to differences in the illuminated

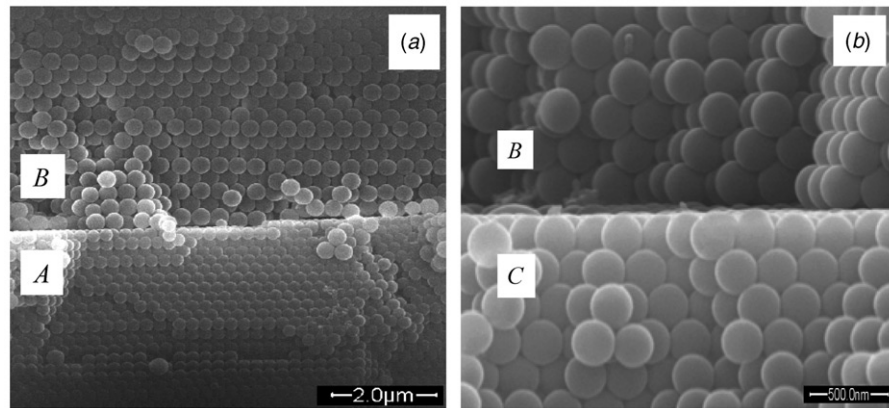


Figure 5. The cross-section SEM images of (a) the AB structure and (b) the CB structure.

region for the two cases. The transmission spectra measured from the bottom (dashed line) and the top (solid line) of this structure are displayed in Figure 4(b). Similarly, the change found in the transmission modes measured in both directions is also less than that in the reflection modes, demonstrating the smaller influence of the imperfections in the sample on the transmission spectrum. A shallower PBG in the transmission spectrum measured from the bottom of the sample, as compared with that measured from the top, also demonstrates the upper crystal (B) with more disorder inside it. If the normal incidence reflection spectrum can be measured by other advanced techniques, the attenuation (from both scattering and absorption) exhibited by each heterostructure in both directions can then be quantified by subtracting its reflectance and transmittance from unity.

Interestingly, only a PBG in the transmission spectra of the CB structure is observed in Figure 4(b). In order to investigate the unique optical property of this structure, we fabricated two crystals C and B. The thickness of each individual crystal C and B is 19 layers, equal to the thickness of the corresponding constituent PC in the CB structure. The transmission spectrum of the CB structure measured from the top of the sample is presented in Figure 4(c) (Exp). The reference transmission spectra from each individual crystals C and B are also shown here for comparison. The relative width ($\Delta\lambda/\lambda$) of the PBG in the [111] direction changes from $\sim 11\text{--}13\%$ for the single crystals to $\sim 19\%$ for the CB structure which suggests that the effective band width can be increased by overlapping two kinds of crystal with similar sphere sizes. A sum of the spectra of the individual crystal is also shown in Figure 4(c) (sum) for comparison with the measured spectrum of the heterostructure. Obviously, the spectrum of the heterostructure is similar to, but not identical to, the sum of the spectra of the two individual crystals. In order to obtain a deep insight

into the broader PBG, we have performed numerical simulations for the individual crystals and CB structure by the finite-difference time-domain (FDTD) technique [13,16,17]. The thicknesses of the individual crystals and the two constituent PCs are all 19 layers. The results are presented in Figure 4(d). Different from the measured results, the PBG width of the CB structure is just equal to the sum of the PBG width of the individual crystal. Therefore, the broader PBG is formed, presumably as a result of coherent interference effects between the PBGs of the two crystals owing to the disorder in heterostructure and the roughness at the interface. Therefore, the binary PC heterostructures can exhibit not only double PBGs but also a single PBG by manipulating the lattice constants of the two constituent PCs in the heterostructures. The broader phenomenon suggests the possibility of tailoring mid-gap resonances in PCs. This may be a viable strategy for creating lightweight and large area optical filters.

Cross-section SEM images of these two types of heterostructures are shown in Figure 5. Clearly, in Figure 5(a) the interface between crystal A and B is smooth and well defined. The two crystal regions in the AB structure remains distinct as the integrity of the large particle interface layer is easier to maintain when the particles below are smaller. The particles of both crystal A and B are stacked layer by layer. These demonstrate the high degree of order of the AB structures along the [111] orientation, perpendicular to the substrate. But in contrast to crystal A in the AB structure, much more disorder and defects in crystal B are clearly seen. This is why the intensity of the reflected peaks changes when illuminating along opposite directions. In Figure 5(b), a smooth and uniform interface between crystal C and crystal B is clearly seen. The particles of both crystal C and B are also stacked layer by layer. These also demonstrate the good quality of CB structures. In a word, the quality of

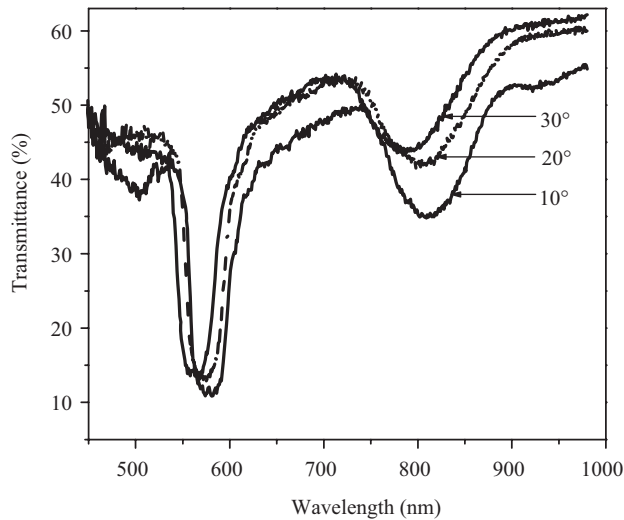


Figure 6. Angle resolved transmission spectra for an AB structure at 10°, 20° and 30° from the normal incidence.

the constituent PCs was largely improved and the roughness at the interface was modified successfully when bigger particles were deposited on top of small particles by the modified self-assembly method.

Before summarizing our research work, we show the dependence of the transmission spectra on the incident angle for the AB structure, as shown in Figure 6. As expected from the Bragg's law $\lambda = 2(2/3)^{1/2}D(n^2 - \sin^2\alpha)^{1/2}$ (where n is the mean refractive index of the structure, D the diameter of the spherical particles, and α is the angle between the normal to the surface and the incident wave) [9], the two PBGs shift to shorter wavelength with increasing incident angle. Meanwhile, a reduction of PBG depth is found for both PBGs. Obviously, this reduction occurs more rapidly in the PBG corresponding to crystal B. Both PBGs remain symmetric even at an incident angle of 30°, implying the high quality of the heterostructure.

4. Conclusions

In summary, we have fabricated two types of heterostructures from PS colloid spheres using the modified self-assembly method and demonstrated the structural and optical quality of the heterostructures with two-layer structures. As compared with samples fabricated previously, the quality of the constituent PCs was largely improved and the roughness at the interface was modified successfully when bigger particles were directly deposited on top of small particles. These binary heterostructures fabricated here exhibit approximately PBG properties, e.g. deep double PBGs or a

broader PBG. Deep double PBGs with steep PBEs show good ordering of the heterostructures fabricated by the modified self-assembly method and provide a strategy for studying late-model ultra-fast all-optical switches. A broader PBG produced by manipulating the sphere sizes of the two constituent PCs in the heterostructures may provide a probability for producing lightweight and large area optical filters. It is possible to form a high quality multilayer heterostructure consisting of many alternating layers of two or more different sphere sizes using the modified self-assembly method. The optical properties of such heterostructures would present an entirely different way of adjusting the photonic properties of these PCs.

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